

Relationship between resistivity and specific heat in a canonical non-magnetic heavy fermion alloy system: $\text{UPt}_{5-x}\text{Au}_x$

B. Andraka^a, R. Pietri^a, S. G. Thomas^a, G. R. Stewart^{a,b},
E. - W. Scheidt^b, and T. Schreiner^b

^a*Department of Physics, University of Florida,
P.O. Box 118440, Gainesville, Florida 32611*

^b*University of Augsburg, Memminger Str. 6, 86135 Augsburg, Germany*

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$\text{UPt}_{5-x}\text{Au}_x$ alloys form in a single crystal structure, cubic AuBe_5 -type, over a wide range of concentrations from $x = 0$ to at least $x = 2.5$. All investigated alloys, with an exception for $x = 2.5$, were non-magnetic. Their electronic specific heat coefficient γ varies from about 60 ($x = 2$) to about 700 mJ/mol K^2 ($x = 1$). The electrical resistivity for all alloys has a Fermi-liquid-like temperature variation, $\rho = \rho_0 + AT^2$, in the limit of $T \rightarrow 0$ K. The coefficient A is strongly enhanced in the heavy fermion regime in comparison with normal and transition metals. It changes from about 0.01 ($x = 0$) to over 2 $\mu\Omega \text{ cm/K}^2$ ($x = 1$). A/γ^2 , which has been postulated to have a universal value for heavy fermions, varies from about 10^{-6} ($x = 0, 0.5$) to $10^{-5} \mu\Omega \text{ cm (mol K/mJ)}^2$ ($x > 1.1$), thus from a value typical of transition metals to that found for some other heavy fermion metals. This ratio is unaffected, or only weakly affected, by chemical or crystallographic disorder. It correlates with the paramagnetic Curie-Weiss temperature of the high temperature magnetic susceptibility.

71.27.+a, 71.10.Ay, 75.30.Mb

Despite more than two decades of research on heavy fermions, the understanding of the phenomena remains unsatisfactory, especially for actinide-based systems. A notable advance in delineating the origin of the enhanced electronic mass in Ce-based systems has been achieved via alloying studies. [1] Such studies are able to show correlations between various low temperature characteristics of materials within a single crystal structure. Few successful alloying studies involving U-based heavy fermions have been reported so far. One of the serious obstacles in such investigations is the proximity to magnetism and the robust character of magnetism in U-compounds and alloys. Thus, an ideal candidate for such an investigation should clearly possess a ground state that is neither magnetic nor superconducting. To avoid any complications due to anisotropy of the electronic properties associated with the crystal structure, materials crystallizing in cubic structures are preferred.

There are only few U-alloys and compounds that can be undoubtedly classified as heavy fermions and which satisfy these prerequisites. One of them, and probably the clearest case is UPt_4Au . [2] This alloy forms in a cubic AuBe_5 -type crystal structure. Its low temperature specific heat divided by temperature is almost 700 mJ/mol K^2 at 1 K. The ratio of the magnetic susceptibility to the specific heat coefficient at low temperatures and the Wilson ratio are small in comparison with all other known heavy fermion systems. Equally small ratios have been found only for two heavy fermion superconductors, UPt_3 and UBe_{13} [3] which lead to speculations that UPt_4Au can be a heavy fermion superconductor at sufficiently low temperatures. However, magnetization measurements [4]

performed down to 20 mK have not yielded any evidence of superconductivity or magnetism.

It has been reported [5] that $\text{UPt}_{5-x}\text{Au}_x$ alloys form a single-phase, cubic AuBe_5 -crystal structure over an extended range of concentrations x ($x = 0$ to at least $x = 2.5$). At the same time, their electronic properties, like the low temperature specific heat and magnetic susceptibility, vary a great deal, making this pseudobinary alloy system a unique and convenient system to study the development of a heavy fermion state upon varying x , and to search for possible deviations from Landau's Fermi-liquid theory in the heavy-fermion regime.

$\text{UPt}_{5-x}\text{Au}_x$ alloys have been prepared by arc melting. All alloys in the range $x = 0$ to 2.5 were single phase according to the X-ray diffraction analysis. Based on a previous study, [5] which did not find any significant dependence of the low temperature properties of these alloys on annealing, we have not annealed our samples. In our present investigation, we have used some of those previously investigated unannealed samples.

Specific heat has been measured between about 1 K and 10 K with the exception for UPt_4Au , which has been studied to 0.35 K. The total specific heat divided by temperature (C/T) versus temperature square is shown in Fig. 1 for eight different alloys corresponding to x between 0 and 2.5. C/T at the lowest measured temperature, which we call further γ , spans a large range of values. It increases from about 80 mJ/mol K^2 for $x = 0$ to almost 700 mJ/mol K^2 for $x = 1$, and then drops again to about 60 mJ/mol K^2 for $x = 2$, thus changing by more than an order of magnitude.

A better estimate of γ would be C/T at 0 K obtained

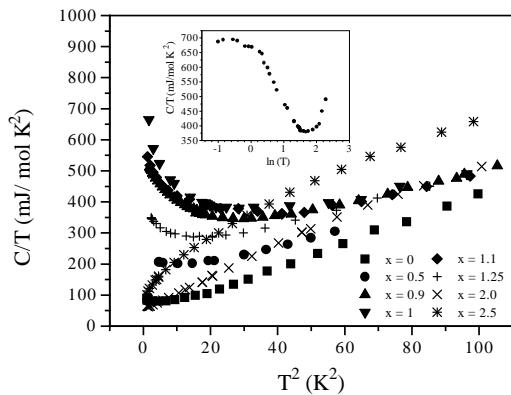


FIG. 1. C/T versus T^2 for $UPt_{5-x}Au_x$ alloys. The inset shows C/T versus $\ln T$ for $x=1$ and temperatures between 0.3 and 10 K.

from the fit of the low temperature specific heat data to some theoretical function. This could be especially important for UPt_4Au which exhibits a number of similarities to extensively studied UCu_4Pd [6,7]. In this later compound, also forming in the $AuBe_5$ -type crystal structure, C/T diverges as T approaches zero. C/T of UPt_4Au , between about 1 and 10 K, has also a quasi-logarithmic temperature dependence if the phonon contribution is subtracted. However, below 1 K, the C/T data show a clear tendency towards saturation (and possibly a very weak maximum near 0.6 K which can not be resolved due to the data scattering). This saturation is shown in the inset to Fig. 1 by expressing the data in the C/T versus $\ln T$ format. C/T between 1.1 and 0.36 K varies by less than 5 % which is less than the absolute accuracy of the measurement, about 10 %. This result provides some justification of associating γ with the value of C/T at about 1 K.

In agreement with Ref. 8, we have found that the low temperature specific heat data of UPt_5 can be well described by a spin-fluctuation formula. This formula can still be applied to $x = 0.5$, but fails to describe the temperature dependence of C/T for all other investigated concentrations. The specific heat for these other concentrations can not be associated with either Kondo-type behavior or, more generally, with any type of behavior corresponding to a single-impurity scaling, $C(T, T_K) = C(T/T_K)$. [9] Because of almost identical molecular weights across the series, a small phonon contribution at 1 K is expected to be constant. Attempting to account for the phonon contribution, we have investigated two non- f $AuBe_5$ homologues: $ThPt_4Au$ and YPt_4Au . [10] The specific heat for both $ThPt_4Au$ and YPt_4Au has a spin-fluctuation temperature dependence

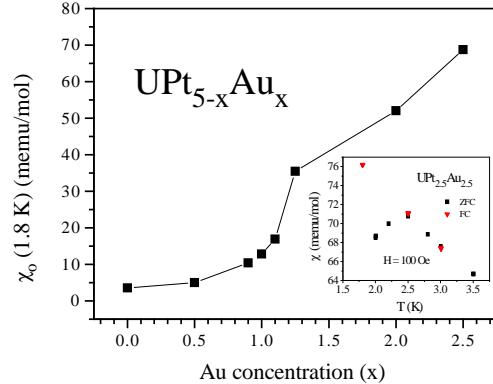


FIG. 2. χ_o versus x for $UPt_{5-x}Au_x$. The inset shows zero-field-cooled (ZFC) and field-cooled (FC) susceptibility ($H = 100$ Oe) for $UPt_{2.5}Au_{2.5}$.

with γ values of 11 and 17 mJ/mol K², respectively. Since neither of them contains 5 f -elements, the spin-fluctuations have to be associated with 5 d -electrons. This is an important observation suggesting that the 5 d electrons of Pt can also be responsible for the spin-fluctuation term in the specific heat of UPt_5 , and that 5 f -5 d hybridization effects can play a significant role in other $UPt_{5-x}Au_x$ alloys, especially in those corresponding to small values of x . A similar conclusion has been reached by authors of Ref. 8, based on a magnetic susceptibility study. They have argued that an unusually large absolute value of the Curie-Weiss temperature (~ 500 K) in the magnetic susceptibility of the pure UPt_5 compound reflects strong 5 f -delocalization effects due to the 5 d -electrons of Pt.

Magnetic susceptibility at the lowest temperature measured, χ_o (1.8 K), shown as a function of x in Fig. 2, does not correlate with γ at all. χ_o increases monotonically from 3.6 to almost 70 memu/mol between $x = 0$ and $x = 2.5$ (Fig. 2). Thus the ratio χ_o/γ is not constant across the system, as expected from the single-impurity model, but increases with x . The especially strong increase in χ_o for $x > 1.25$ prompted us to perform low field magnetization measurements on these alloys. These measurements for $UPt_{2.5}Au_{2.5}$ revealed a maximum at about 2.5 K (T_M) in the zero field cooled susceptibility (ZFC) and a discrepancy between zero-field-cooled and field-cooled (FC) susceptibilities at low temperatures (Inset to Fig. 2). No corresponding maximum has been found in the specific heat data. Similar spin-glass-like anomalies have been observed in other γ -enhanced U- or Ce-based systems, with chemical formulas of the form $M(A_xB_{1-x})$, near $x = 0.5$; where M is either U or Ce, and A and B are normal or transition metals. [11] Although

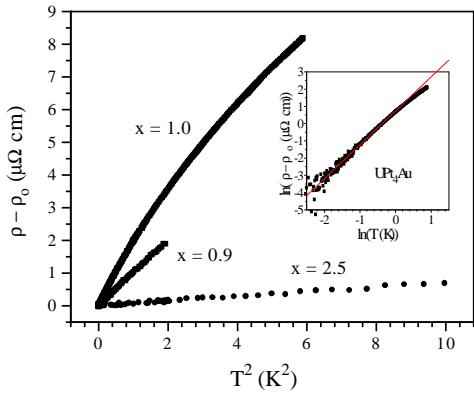


FIG. 3. $\rho - \rho_0$ versus T^2 for $x = 0.9, 1$, and 1.25 . The inset shows the zero-temperature limit of the resistivity for UPt_4Au on a logarithmic scale.

there is no consensus as to the nature of the ground state (or different ground states) in these alloys, it is believed that non-magnetic atom disorder (NMAD) is responsible for their spin-glass-like properties. Low field magnetization measurements performed on all other alloys, including UPt_2Au_3 , have not detected any anomalies, although small discrepancies (up to 5%) between ZFC and FC susceptibility have been found for $x = 2$ below 3 K. The maximum value of T_M occurring for $x \simeq 2.5$ in the $\text{UPt}_{5-x}\text{Au}_x$ system is consistent with NMAD reaching its maximum near $x = 2.5$. Magnetic measurements indicate further that $\text{UPt}_{5-x}\text{Au}_x$ alloys are not completely free from complications associated with the nearness to magnetism. Nevertheless, such complications can be safely ruled out in the range of x where γ undergoes especially spectacular changes ($0 \leq x \leq 1.25$).

Before addressing our resistivity results we point out the important issue of preferential site occupancy. Strong variation of the thermodynamic low temperature properties of $\text{UPt}_{5-x}\text{Au}_x$ near $x=1$ and their extremal values for $x=1$ have been attributed to preferential atomic occupancy of non-U sites [2,5]. Pt would preferentially occupy trigonal symmetry 16e sites while Au would go on cubic 4c sites. Additional, indirect support for this explanation is given by thermodynamic investigations of the aforementioned U-alloy system $\text{UCu}_{5-x}\text{Pd}_x$, existing in the same crystal structure. The concentration corresponding to $x=1$ also exhibits extremal values of its low temperature properties in comparison to other alloys belonging to $\text{UCu}_{5-x}\text{Pd}_x$ system. Note that the smaller atoms (Cu, Pt) would preferentially occupy majority 16e sites, while larger atoms would occupy 4c sites in both alloy systems. Previous attempts using X-ray diffraction and resistivity studies to clarify this issue for $\text{UPt}_{5-x}\text{Au}_x$

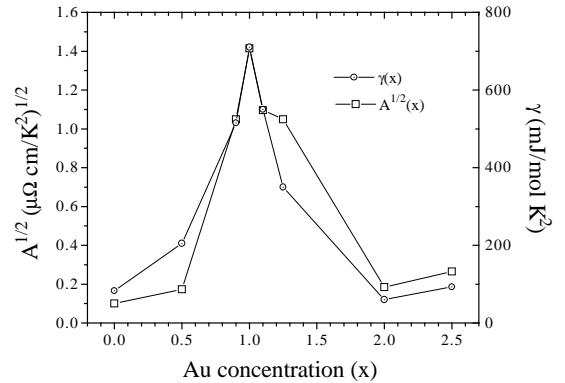


FIG. 4. $A^{1/2}$ and γ versus x for $\text{UPt}_{5-x}\text{Au}_x$.

were inconclusive [5]. However, a recent elastic neutron scattering study [12] on the related $\text{UCu}_{5-x}\text{Pd}_x$ has confirmed the preferential non-U site occupancy. Thus, a similar preferential site occupancy and chemical ordering of $x=1$ is highly probable for $\text{UPt}_{5-x}\text{Au}_x$.

This resistivity for all investigated alloys has a Fermi-liquid-like temperature dependence, $\rho = \rho_0 + AT^2$, at sufficiently low temperatures. This variation is shown in Fig. 3 for $x = 0.9, 1$, and 2.5 in the form of $\rho - \rho_0$ versus T^2 . The residual resistivity ρ_0 , obtained on samples derived from a single batch of U, increases with x from about $25 \mu\Omega \text{ cm}$ for $x = 0$ [8] to about $165 \mu\Omega \text{ cm}$ for $x = 2.5$. Thus in agreement with the previous study, our resistivity results do not directly support the notion of a chemical ordering near $x=1$; i.e., no discernable dip in ρ_0 is observed for this concentration. We have noticed some variation of ρ_0 between samples obtained from U-batches having different chemical purity. One of our UPt_4Au samples had $\rho_0 \simeq 40 \mu\Omega \text{ cm}$, the other one had $\rho_0 \simeq 80 \mu\Omega \text{ cm}$. Interestingly, the temperature square coefficient of the resistivity A , discussed next, was identical for both $x=1$ samples, within the experimental uncertainty (10 %).

The most striking feature of Fig. 3 is a large variation, from concentration to concentration, in the slope of the $\rho - \rho_0$ versus T^2 . This slope corresponds to the A coefficient. In the investigated range of concentrations, A changes, in a non-monotonic fashion, from its lowest value of about $0.01 \mu\Omega \text{ cm/K}^2$ for $x = 0$ to over $2 \mu\Omega \text{ cm/K}^2$ for $x = 1$, thus by a factor larger than 200. The A coefficient fairly well tracks the trends for the changes in γ , as demonstrated in Fig. 4. The format of this figure, $A^{1/2}$ and γ versus x , has been chosen based on the observation of Kadowaki and Woods, [13] that for a large number of heavy fermion compounds $A \simeq 10^{-5} \gamma^2$, where

A is expressed in $\mu\Omega$ cm/K² and γ in mJ/mol K². A similar relationship has been found in transition metals, but with the average ratio A/γ^2 about 20 times smaller than that in heavy fermion compounds. This proportionality between A and γ^2 in transition metals has been accounted for theoretically [14] in terms of Baber's model [15] describing electron-electron scattering. Also, the electron-electron scattering is believed to be the source of the enhanced values of A in heavy fermion systems [16,17]. However, the large ratio A/γ^2 , in comparison with transition metals, has not yet been fully explained. Furthermore, the universality of this ratio has not been established. The ratio of A and γ^2 is not constant across the $\text{UPt}_{5-x}\text{Au}_x$ series (Fig. 5). This ratio is of order $10^{-6} \mu\Omega \text{ cm} (\text{mol K/mJ})^2$ only for the pure UPt_5 compound and the $x = 0.5$ alloy, thus within the range of values corresponding to many transition metals. A/γ^2 grows to $4 \times 10^{-6} \mu\Omega \text{ cm} (\text{mol K/mJ})^2$ for $x = 0.9, 1$, and 1.1 , and reaches a value close to the claimed universal ratio for heavy fermion compounds for $x = 1.25, 2$, and 2.5 . Our results confirm the correlation between A and γ^2 , but they also indicate that the ratio of these two quantities can vary by more than a decade, in disagreement with recent theoretical predictions [17]. To the best of our knowledge, this is the first observation of such a correlation in an alloy system over a large range of concentrations, as opposed to a previous study on pure compounds possessing full translational symmetry. [13]

It has been postulated [16] that disorder can influence A/γ^2 values. Both, the ratio A/γ^2 and the amount of crystallographic disorder, in general, increase with x in our alloy system, therefore implying that the disorder increases this ratio. However, such a possibility is unlikely considering that the two UPt_4Au samples studied showed significantly different residual resistivities, as discussed earlier, and yet had identical values of A/γ^2 within the accuracy of our measurement. Thus crystallographic or atomic disorder seems to play a rather minor, if any, role in the evolution of A/γ^2 throughout the series.

It has been also argued, in the framework of the spin-fluctuation model, [17] that this ratio, which should be almost universal for heavy fermions, can be enhanced in the proximity to a magnetic instability. According to experimental studies [3], the Wilson ratio ($\chi_o/\gamma\mu_{\text{eff}}^2$) can be used as an indication of such a proximity to magnetism. Since μ_{eff} extracted from the high temperature susceptibility is almost constant across this pseudobinary alloy system, one may use the χ_o/γ ratio instead. χ_o/γ versus x is shown in Fig. 5 together with corresponding A/γ^2 values. Although both ratios increase above $x = 1.25$, there is no correlation between these two quantities for $x < 2$. A somewhat better correlation is observed between A/γ^2 and the absolute value of the Curie-Weiss temperature ($|\Theta_{\text{CW}}|$). A decrease of $|\Theta_{\text{CW}}|$ corresponds to an increase of A/γ^2 . In particular, $|\Theta_{\text{CW}}|$ and A/γ^2 are constant for $x = 0.9, 1$, and 1.1 , thus for a concen-

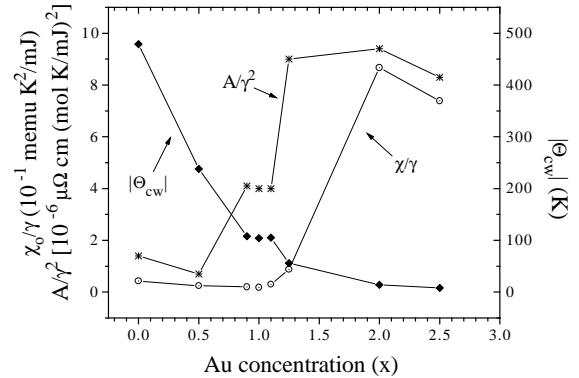


FIG. 5. A/γ^2 , χ_o/γ , and $|\Theta_{\text{CW}}|$ versus x .

tration range corresponding to large changes of γ . It is reasonable to associate large values of $|\Theta_{\text{CW}}|$ in this alloy system, in agreement with Ref. 6 and also with our specific heat data, with strong $5f$ - $5d$ hybridization, and consequently with a large degree of delocalization of $5f$ electrons. Alloys for which we expect especially strong $5f$ - $5d$ hybridization, as evidenced by large paramagnetic Curie-Weiss temperatures ($x \sim 0$), have A/γ^2 values in the range corresponding to transition metals. Upon a decrease of $|\Theta_{\text{CW}}|$, A/γ^2 increases and reaches a value close to $10^{-5} \mu\Omega \text{ cm} (\text{mol K/mJ})^2$, found in many other heavy fermion systems.

The discussed Fermi-liquid-like temperature dependence in the resistivity of $\text{UPt}_{5-x}\text{Au}_x$ alloys is observed only at the lowest temperatures, and the range of temperatures in which it holds varies with x . The upper temperature limit of this variation seems to correlate with the inverse of γ ; e.g., it is about 1 K for $x = 0.9, 0.7$ K for $x = 1.0$, 4 K for $x = 2.0$, and about 3 K for $x = 2.5$. This observation is rather surprising since it is generally believed that the $\rho = \rho_o + AT^2$ temperature dependence is a property of the so-called coherence regime, which is a consequence of a translational periodicity of the system. Our results do not preclude the possibility that replacing Pt by Au, leading to the destruction of this symmetry, affects somehow the upper temperature limit of the discussed temperature variation, but it does not seem to be a dominating cause.

Finally, our results have implications on the possible explanation of the origin of the non-fermi-liquid-like low temperature properties for the extensively studied $\text{UCu}_{5-x}\text{Pd}_x$ alloys [6,7]. One of the leading explanations [18] is based on the Kondo-disorder idea [19]. Despite the obvious ligand disorder also in $\text{UPt}_{5-x}\text{Au}_x$ alloys, their properties stay fermi-liquid-like over a wide range

of x . This observation imposes severe limitations on the applicability of the Kondo-disorder theory to UCu_4Pd .

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